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#### **REGULAR PAPER**



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# Towards the partial resumption of agriculture with buckwheat cultivation in fields physically decontaminated of radioactive cesium after the nuclear power plant accident in 2011: a case study in Yamakiya District, Fukushima

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#### ABSTRACT

Yamakiya District in the town of Kawamata, Fukushima Prefecture, was evacuated after the nuclear accident at the Tokyo Electric Power Company's Fukushima Dai-ichi nuclear power plant in 2011. Since then, nuclear decontamination procedures have been applied to the surrounding environment, including agricultural fields. The decontamination procedure for agricultural fields consists of the removal of radiation-contaminated surface soil, followed by soil dressing and plowing. However, radioactive cesium (RCs) remains in the soil even after decontamination. In this study, we investigated the effect of applying nitrogen, potassium fertilizers, and cattle manure compost on buckwheat growth and the concentration of RCs in the grain of buckwheat cultivated in a decontaminated field from 2014 to 2016. Applications of potassium fertilizer and cattle manure compost increased the soil exchangeable potassium content and decreased the RCs concentration in the grain of buckwheat cultivated in the decontaminated field. Before the cultivation of the first buckwheat crop, the RCs concentration in soil varied widely and there were 'hot spots' with high RCs concentrations because of insufficient mixing of the original and the dressed soils. Therefore, soil had to be adequately mixed to avoid producing grain with a high RCs concentration. Buckwheat grew better when supplied with more nitrogen fertilizer than the conventional amount at the first cultivation, indicating that the dressed soil had low fertility. We also monitored buckwheat cultivation by local farmers in decontaminated fields from 2015 to 2017. By using potassium fertilizer, the farmers produced buckwheat grain with low RCs concentrations from 2015 to 2017.

#### 1. Introduction

An accident at the Tokyo Electric Power Company's (TEPCO) Fukushima Dai-ichi (No. 1) nuclear power plant (FDNPP) triggered by the Great East Japan Earthquake and tsunami on 11 March 2011 resulted in the emission of radioactive materials, including radioactive cesium ( $^{134}Cs + ^{137}Cs$ , RCs), into the environment of eastern Japan (Fukushima Prefecture, 2015). Evacuation zones were established in this area to reduce radiation exposure to the residents. Such zones occupied 371 km<sup>2</sup> in Fukushima in December 2017 (Fukushima Prefecture, 2017a), although the total area of evacuation zones has since reduced because of the decontamination of radioactive materials in the environment and natural attenuation of radiation. In October 2017, there were still 20,000 evacuees from evacuation zones (Fukushima Prefecture, 2017a).

Before the accident at the FDNPP, 217 farmer households in Yamakiya District, Kawamata town (Figure 1) **ARTICLE HISTORY** 

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soil fertility

conducted agricultural activities across approximately 880 ha of farmland (including paddy fields, upland fields, and pastures) (Kawamata town, 2013). After the accident, farmland with RCs concentrations in soil up to 5000 Bq kg<sup>-1</sup> was physically decontaminated. This process involved the removal of radiation-contaminated surface soil (to about 5-cm soil depth), soil dressing, and plowing to mix the new soil into the original soil and reduce the RCs concentration (MAFF, 2013; Yukumoto, 2012). Approximately 600 ha of farmland was targeted for physical decontamination in Yamakiya District (MEV, 2017). Due to the decrease in the RCs concentrations as a result of physical decontamination of residential land, buildings, roads, forests, and farmland, as well as by natural attenuation, the evacuation order for Yamakiya was lifted in March 2017 (Nuclear Emergency Response Headquarters, 2016).

In Yamakiya District, buckwheat (*Fagopyrum esculentum* Moench) was richly cultivated before the accident at the FDNPP ('Nihon Keizai Shimbun' on 30 December 2011). Some farmers intended to relaunch agriculture by

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Supplemental data for this article can be accessed here.

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**Figure 1.** Location of Yamakiya and RCs deposition maps. (a) 1 November 2011 and (b) 4 November 2015. Cited from 'Extension Site of the Distribution Map for Radiation Dose' (http://ramap.jmc.or.jp/map/eng/).

cultivating buckwheat in physically decontaminated fields in 2015. Buckwheat would be a suitable crop to cultivate in such fields in term of adaptation to low-fertile soil condition (Pavek, 2016) because the dressed soil added during physical decontamination may have low fertility, as reported by Yoshino et al. (2015). Previously, we have shown that the RCs concentration in buckwheat grain can be decreased by increasing the soil exchangeable potassium (ExK) content in RCs-contaminated fields which were not conducted the physical decontamination (Kubo et al., 2015). Recently, Kubo et al. (2017) have also reported that increased soil ExK content affects to decreased RCs absorption due to the decreased soil exchangeable RCs concentration and to decreased RCs translocation from root to shoot and from shoot to grain. In addition, Saito and Sato (2014) have reported that the application of cattle manure is useful to increase the soil ExK content and to decrease the RCs concentration in cabbage. At that time, however, there was insufficient information about the soil RCs concentrations in physically decontaminated fields, and about the effect of soil ExK content to RCs concentrations in the grain of buckwheat grown in those fields. In this study, therefore, we investigated the soil RCs concentration and the effect of soil ExK content on grain RCs concentrations in buckwheat grown in physically decontaminated fields in Yamakiya District. We also evaluated the effects of increased nitrogen application rates and cattle manure application on buckwheat growth and soil fertility. Besides the on-site experiments mentioned above, we monitored the resumption of buckwheat cultivation by local farmers in fields that had been physically decontaminated of RCs.

#### 2. Materials and methods

#### 2.1. Field experiment

A field experiment was conducted at a farmer's field during 2014–2016 in Yamakiya, where physical decontamination of RCs had been conducted in the spring of 2014 by the Ministry of the Environment (MEV). The <sup>137</sup>Cs concentration in the physically decontaminated field was

1236  $\pm$  84 Bq kg<sup>-1</sup> (average  $\pm$  standard error, n = 15) before sowing in 2014. Before buckwheat cultivation, N (ammonium sulfate, 21.0% N) and P<sub>2</sub>O<sub>5</sub> (calcium superphosphate, 20.5%  $P_2O_5$ ) were each applied at 30 kg ha<sup>-1</sup> uniformly across the field. This is the conventional rate of application for buckwheat cultivation (Fukushima prefecture, 2017b). K<sub>2</sub>O (potassium sulfate, 50.0% K<sub>2</sub>O) was applied at four rates: 0, 200, 400, and 600 kg ha<sup>-1</sup>, corresponding to 150, 250, 350, and 450 mg  $K_2O$  kg<sup>-1</sup> soil ExK content, respectively (calculated as the soil bulk density 1.0 mg m<sup>-3</sup>, soil depth 20 cm), in 2014. The plots supplemented with  $K_2O$  at 0, 200, 400, and 600 kg ha<sup>-1</sup> were designated as N1K1, N1K2, N1K3, and N1K4, respectively (Table 1). The soil ExK content in the N1K1 plot before sowing was 180 and 200 mg  $K_2O$  kg<sup>-1</sup> in 2015 and 2016, respectively. In addition, a plot with N applied at 60 kg ha<sup>-1</sup> was established in the N2K3 plot (400 kg  $ha^{-1}$  K<sub>2</sub> O application). In 2016, two plots were added: 10 t  $ha^{-1}$ of cattle manure compost and no manure compost (M1 and M0 plots, respectively) in an area with no cropping history since 2011 in the same field. The N, P<sub>2</sub>O<sub>5</sub>, and K<sub>2</sub> O fertilizers were each applied at a rate of 30 kg  $ha^{-1}$  in the M1 and M0 plots. All fertilizers and manure were applied before plowing and sowing. The area of each plot was 18.0 m<sup>2</sup> for the N1K1, N1K2, N1K3, N1K4, and N2K3 plots and 26.25 m<sup>2</sup> for the M0 and M1 plots. The buckwheat cultivar 'Aizunokaori', which was bred in Fukushima Prefecture (Yamauchi, 2012), was sown after hill plowing. The sowing dates were 5 August, 30 July, and 28 July in 2014, 2015, and 2016, respectively. The seed was sown by row seeding with the row space of 0.75 m (seeding rate, 50 kg ha<sup>-1</sup>). The experiment had a randomized block design with three replications for the N1K1, N1K2, N1K3, N1K4, and N2K3 plots and two replications for the M0 and M1 plots. The field was surrounded by an electric fence to avoid crop damage by wild animals such as boars.

The shoot and grain of buckwheat were sampled by cutting at ground level at the maturity stage (2 October, 8 October, and 5 October in 2014, 2015, and 2016, respectively). The grain was separated from the shoot by threshing, and was polished using a polishing

Table 1. Amount of N,  $K_2O$ , and cattle manure compost applied to each plot in the field experiment.

	2014–2015		2016		
	Ν	K <sub>2</sub> O	Ν	K <sub>2</sub> O	compost
	(kg ha <sup>-1</sup> )	(t ha <sup>-1</sup> )			
N1K1	30	0	30	0	-
N1K2	30	200	30	200	-
N1K3	30	400	30	400	-
N1K4	30	600	30	600	-
N2K3	60	400	60	400	-
MO	_	_	30	30	0
M1	_	-	30	30	10

machine (F1; Fukubishi Co., Ltd., Fukushima, Japan) to avoid contamination of the grain surface (Kubo et al., 2016). Soil samples at 20-cm depth were collected from around the plant root with a worm scoop (Fujiwara Scientific Company Co., Ltd., Tokyo Japan) during plant sampling. Ten soil samples were collected using the scoop in each plot and then mixed before further analysis.

### **2.2.** Buckwheat cultivation by local farmers in RCs decontaminated fields

In 2015, buckwheat was cultivated by a local farmers' group, the 'Kawamata-Nakanouchi Buckwheat Club (KNBC)' (Supplementary Figure 1a), in three fields (fields A (22 a), B (20 a), and C (33 a)) in Yamakiya District, which had been physically decontaminated of RCs by MEV. First, we explained to the members of the KNBC about the use of potassium fertilizer to decrease the RCs concentration in buckwheat, as demonstrated previously (MAFF, 2014; Kubo et al., 2015). Before buckwheat sowing, soil samples were collected from 0 to 20 cm depth at five sites in each field, and these samples were used to measure the RCs concentration and soil ExK content. Based on the lowest soil ExK content in each field, potassium sulfate was added to increase the soil ExK content to 300 mg K<sub>2</sub>O kg<sup>-1</sup> (Kubo et al., 2015). The amount of potassium sulfate applied was 580, 588, and 900 kg  $ha^{-1}$  in fields A, B, and C, respectively. The N and P2O5 fertilizers were added at a rate of 60 kg ha<sup>-1</sup> (based on the results of '1. Field experiment') and 30 kg ha<sup>-1</sup>, respectively. On 8 August, fertilization, plowing, and sowing by broadcasting were conducted. The local buckwheat variety 'Yamakiyazairai' was sown at a density of 50 kg seeds ha<sup>-1</sup>. On 18 October, buckwheat was harvested from all fields by GC360 combine harvester (Yanmar, Japan) а (Supplementary Figure 1b). The grain was passed through a winnower and polished. Soil samples were collected at 0-20-cm depth from five sites in each field at the seedling, flowering, and maturity stages of buckwheat.

Buckwheat was cultivated by the members of the KNBC in four fields (total, 1.2 ha) and five fields (total, 1.5 ha) in Yamakiya District in 2016 and 2017, respectively. In 2016 and 2017, the soil ExK content at the harvest in previous years in each field was determined as the average value of five sites, and K<sub>2</sub>O was added to increase the soil ExK content to 300 mg K<sub>2</sub>O Kg<sup>-1</sup>. In 2016, all the fields had up to 300 mg K<sub>2</sub>O Kg<sup>-1</sup> of soil ExK content. In 2017, the soil ExK content of one field (Field B) did not exceed 300 mg K<sub>2</sub>O Kg<sup>-1</sup> (251 mg K<sub>2</sub> O Kg<sup>-1</sup>). The amount of potassium sulfate application

was 60 kg ha<sup>-1</sup> and 60–196 kg ha<sup>-1</sup> in 2016 and 2017, respectively (the fields with soil containing 300 mg K<sub>2</sub> O kg<sup>-1</sup> at harvest in previous years were supplemented with 30 kg K<sub>2</sub>O ha<sup>-1</sup>, the conventional application rate). The N and P<sub>2</sub>O<sub>5</sub> fertilizers were each added at a rate of 30 kg ha<sup>-1</sup> in both years. Fertilization, plowing, and sowing were conducted on 31 July and 5 August in 2016 and 2017, respectively, and harvesting was conducted on 30 September and 28 September in 2016 and 2017, respectively. The buckwheat variety, sowing method and density, and harvesting and preparation after the harvest were same as those in 2015.

#### 2.3. Soil and plant analyses

Soil samples were air-dried and passed through a 2.0-mm sieve to determine the soil ExK content and the RCs concentration. The water content in the air-dried soil samples in '2.1 Field experiment' was 2.7 ± 0.2%, 2.5  $\pm$  0.1%, and 2.7  $\pm$  0.1% (average $\pm$  standard error in all the samples) in 2014, 2015, and 2016, respectively. The soil ExK content was determined by atomic absorption spectrophotometry (ZA-3000, Hitachi High-Technologies Corporation, Tokyo, Japan) after extraction in 1 M ammonium acetate at a soil solution ratio of 1:10 for 1 h at room temperature. The other chemical properties (pH, exchangeable CaO, exchangeable MgO, cation exchange capacity (CEC), available P<sub>2</sub>O<sub>5</sub>, NH<sub>4</sub>-N, NO<sub>3</sub>-N, P<sub>2</sub> O<sub>5</sub> absorption coefficient, and humus) of soil in the N1K1 plots at sowing and maturity each year were analyzed based on the methods described by Kubo et al. (2018). Analyses of the soil chemical properties except for ExK content were conducted for a bulk soil sample, which was a mixed sample of three replicates collected at each sampling time. Bulk soils of M0 and M1 at the maturity stage of buckwheat were also analyzed to determine their chemical properties (including soil ExK content).

In '2.1 Field experiment', grain yield was measured for each plot with 15% water content. In '2.2. Buckwheat production by local farmers in RCs-decontaminated fields', the total weight of grain harvested from all the fields was recorded. For the RCs analysis, we targeted only <sup>137</sup>Cs because it is difficult to measure <sup>134</sup>Cs accurately due to natural attenuation (half-lives; <sup>134</sup>Cs 2.0648 year, <sup>137</sup>Cs 30.1671 year). The ratio of <sup>134</sup>Cs and <sup>137</sup>Cs emitted from TEPCO's FDNPP was approximately 1:1 (Komori et al., 2013). To evaluate the RCs concentration in buckwheat grain produced by the KNBC, the <sup>134</sup>Cs concentration was estimated from the <sup>137</sup>Cs concentration and the half-lives of <sup>134</sup>Cs and <sup>137</sup>Cs. The <sup>137</sup>Cs concentration was measured with a germanium semiconductor detector (GC2520-7500SL; Canberra Japan KK, Tokyo, Japan) in NARO for airdried all soil samples and for buckwheat grain (15% water content) at the maturity stage for all the samples of '2.1 Field experiment' and the 2015 and 2016 samples of '2.2. Buckwheat production by local farmers in RCs-decontaminated fields'. Buckwheat grain produced by the KNBC in 2017 was measured with a germanium semiconductor detector in Fukushima prefecture. The measurement of the <sup>137</sup>Cs concentration had a 10% range error, and the results were decay-corrected to the day of sampling. The transfer factor (TF) of RCs from soil to buckwheat grain was calculated as follows: TF = <sup>137</sup>Cs (Bq kg<sup>-1</sup>) in buckwheat grain (15% water content)/<sup>137</sup>Cs (Bq kg<sup>-1</sup>) in air-dried soil.

#### 2.4. Statistical analyses

To detect differences among treatments, data were subjected to analysis of variance (ANOVA) using the general linear model procedure, and multiple comparisons were conducted with the least significant difference (LSD) procedure. Pearson's correlation analyses were used to evaluate the relationships between two characteristics. Analyses were conducted using SPSS software (IBM SPSS ver. 25 for Windows; IBM Japan).

#### 3. Results and discussion

#### 3.1. Field experiment

#### 3.1.1. Plant growth and grain yield

The average shoot dry weight (DW) of buckwheat in all plots was 1314 and 2216 kg ha<sup>-1</sup> in 2014 and 2015, respectively, and was greater in 2015 than in 2014 (Table 2). In 2014, the shoot DW differed significantly among plots, and was higher in N2K3 than in the other plots. The shoot DW did not differ significantly among the plots in 2015. The average grain yield of buckwheat from all plots was 966, 1022, and 898 kg ha<sup>-1</sup> in 2014, 2015, and 2016, respectively. The grain yield from the N2K3 plot tended to be higher than those from the other plots in 2014, although the differences among plots were not significant. The growth of buckwheat in areas without fertilizer was very poor, and no yield was obtained (data not shown). In 2015 and 2016, the grain yields did not differ among plots.

The CEC, NO<sub>3</sub>-N content, and humus content at sowing were lower in 2014 than in 2015 and 2016. These differences may explain the difference in plant growth among plots in 2014. The NO<sub>3</sub>-N content was low (1.1 mg kg<sup>-1</sup>) in the dressed soil (decomposed granite soil) in the fields (Table 3), similar to the values for dressed soil reported previously by Yoshino et al. (2015). It was considered that the N fertilization was important to increase buckwheat yield as reported by Sugimoto (2004) and Wang et al. (2015). The CEC and humus content were also low,

Table 2. Shoot dry weight (DW), grain yield, and grain <sup>137</sup>Cs concentration in field experiment.

		Shoot DW	Grain yield	Grain <sup>137</sup> Cs
		(kg ha <sup>-1</sup> )	(kg ha <sup>-1</sup> )	(Bq kg <sup>-1</sup> )
2014	N1K1	1395	908	17.0
	N1K2	1141	790	15.8
	N1K3	1220	1015	7.4
	N1K4	1022	612	8.0
	N2K3	1795	1499	8.0
ANOVA		*a	ns	*
LSD (P < 0)	).05) <sup>b</sup>	373	921	6.7
2015	N1K1	2207	1013	14.7
	N1K2	2553	1015	7.0
	N1K3	2085	889	5.9
	N1K4	2262	1131	5.8
	N2K3	1969	1071	5.4
ANOVA		ns	ns	ns
LSD (P < 0)	).05)	809	408	6.4
2016	N1K1	_	939	14.3
	N1K2	-	864	6.2
	N1K3	-	878	5.0
	N1K4	-	796	4.1
	N2K3	-	1015	4.6
ANOVA			ns	**
LSD ( <i>P</i> < 0.05)			417	4.7

<sup>a</sup> \*\* and \* indicate significant difference at P < 0.01 and  $0.01 \le P < 0.05$ , respectively. ns, not significant.

<sup>b</sup> Least significant difference.

4.8 cmol<sub>c</sub> kg<sup>-1</sup> and 0.12%, respectively, in the dressed soil. CEC and humus may also have had interactive effects on buckwheat growth. The conventional basal N fertilizer application rate for buckwheat in Fukushima Prefecture is 0–30 kg ha<sup>-1</sup> (Fukushima Prefecture, 2017b). Therefore, we considered that double the amount of N fertilizer (60 kg ha<sup>-1</sup>) would be suitable to support the first buckwheat crops cultivated in the decontaminated fields in this area.

### 3.1.2. Grain RCs concentration affected by soil ExK content

The range of grain <sup>137</sup>Cs concentrations was 7.4–17.0 Bq kg<sup>-1</sup> in 2014, 5.4–14.7 Bq kg<sup>-1</sup> in 2015, and 4.1–14.3 Bq kg<sup>-1</sup> in 2016 (Table 2), and all these values were under the standard limit (100 Bq kg<sup>-1</sup>) even when the <sup>134</sup>Cs and <sup>137</sup>Cs concentrations were combined. The decrease in

RCs concentration in the field due to the physical decontamination and natural attenuation of RCs would have partly contributed to the lower concentration of RCs in buckwheat grain than the standard limit. The grain <sup>137</sup>Cs concentration differed significantly among plots in 2014 and 2016, respectively, and was significantly higher in the N1K1 plot than in the other plots in both years. Although there was no significant difference in grain <sup>137</sup>Cs concentration among the plots in 2015, it tended to be higher in N1K1 than in the other plots. In 2014, grain in N1K2 had a relatively higher <sup>137</sup>Cs concentration than those of grain in plots with higher application rates of potassium. The grain <sup>137</sup>Cs concentration and <sup>137</sup>Cs TF decreased with higher soil ExK content at maturity in all 3 years (Figure 2), although the relationship between soil ExK content and grain <sup>137</sup>Cs concentration in 2014 was not significant. The <sup>137</sup>Cs TF was stably low when the soil ExK content exceeded 300 mg  $K_2O$  kg<sup>-1</sup>. A decrease in grain RCs concentration with increasing soil ExK content was also observed in a field that had not been physically decontaminated (soil RCs concentration;  $<5000 \text{ Bq kg}^{-1}$ ) (Kubo et al., 2015). Kubo et al. (2017) demonstrated that higher soil ExK content decreased both RCs absorption and RCs translocation (from root to shoot and from shoot to grain) in buckwheat plants. Since some RCs remains in the soil even after physical decontamination, increasing the soil ExK content is required to decrease the RCs concentration in buckwheat grain, whether or not the field has been decontaminated. Increasing the soil ExK content has also been shown to reduce the RCs content in rice (Fujimura et al., 2016). It is important to continuously monitor the RCs transfer from soil to grain in such fields.

### 3.1.3. Effect of variability in soil RCs concentration on grain RCs concentration

As shown in Figure 2, the relationship between soil ExK content and grain <sup>137</sup>Cs concentration was more variable in 2014 than in 2015 and 2016, and the coefficient of correlation was not significant in 2014. However, the coefficient of correlation between soil ExK content and

Table 3. Soil chemical properties in N1K1 plots and dressed soil in field experiment.

	2014		2015		2016		
	Sowing	Maturity	Sowing	Maturity	Sowing	Maturity	Dressed soil
рН	5.6	5.7	5.5	5.8	5.8	5.8	6.4
Exchangeable CaO (mg kg <sup>-1</sup> )	1420	1230	1470	1360	1440	1360	976
Exchangeable MgO (mg kg <sup>-1</sup> )	184	130	178	142	172	146	80.3
Exchangeable $K_2O$ (mg kg <sup>-1</sup> )	152	167	206	116	144	175	49.1
CEC (cmol <sub>c</sub> kg <sup><math>-1</math></sup> )	9.2	9.0	11.4	11.7	11.6	12.7	4.8
Available $P_2O_5$ (mg kg <sup>-1</sup> )	239	214	243	223	193	203	90.7
$NH_4$ -N (mg kg <sup>-1</sup> )	16.3	5.1	20.9	6.4	4.7	4.4	<1.0
$NO_3-N (mg kg^{-1})$	1.1	2.5	4.6	2.0	4.8	2.7	1.1
$P_2O_5$ absorption coefficient (mg kg <sup>-1</sup> )	4850	5380	6050	6110	6440	7150	918
Humus (%)	4.4	4.8	6.1	6.3	5.7	6.7	0.1

Analyses were conducted for bulk soil sample (N1K1 plots: mixture of three replicate samples, dressed soil: mixture of five samples).



Figure 2. Relationships between soil ExK content and grain <sup>137</sup>Cs concentration (left) and <sup>137</sup>Cs transfer factor (right) in field experiment.

Open and closed symbols show the values of each plot and the average of plots, respectively. Correlation coefficients were calculated with power approximation. \*\* and \* show significant at P < 0.1 and 0.1 <=. P < 0.05, respectively. ns shows not significant.

the <sup>137</sup>Cs TF in 2014 was significant and high. The reason for the variation in grain <sup>137</sup>Cs was the variation in soil <sup>137</sup>Cs concentration within the field. The range and coefficient of variation were larger in 2014 than in 2015 and 2016 (Table 4). This result indicated that there was wide variation in the soil <sup>137</sup>Cs concentrations within a field and relatively high RCs concentrations in some localized spots. This may have been due to insufficient mixing of the dressed soil with the original soil, which was not able to be removed completely by the decontamination procedure. Since there is a risk of high RCs concentrations in localized spots in the physically decontaminated fields before cultivation, it is important to carefully mix the original soil and dressed soil by tillage before cultivation and to increase the soil ExK content by adding K fertilizer.

The average soil  $^{137}$ Cs concentration gradually decreased from 2014 to 2016 (Table 4). The effective halflives of soil  $^{137}$ Cs concentrations were 3413 days (from 2014 to 2015), 2373 days (from 2015 to 2016), and 2805 days (from 2014 to 2016) in the field, considerably shorter than the half-life of  $^{137}$ Cs by natural attenuation

**Table 4.** Range, average, and coefficient of variation on the soil<sup>137</sup>Cs concentrations in the field.

	Range	Average	CV
	(Bq kg <sup>-1</sup> )	$\overline{(\text{Bq kg}^{-1})}$	(%)
2014	680-3380	1640	44.8
2015	833-1972	1521	19.4
2016	824–1972	1368	21.1

(*n* = 15).

(11,021 days). MAFF (2017) also has reported that the RCs concentration in the upland fields in Fukushima in November 2015 had decreased by 18% compared with that in November 2014, even though the decrease in RCs by physical attenuation during this period was only 8%. The rapid attenuation of <sup>137</sup>Cs in the upland fields may be related to mixing of surface soil with deeper soil by tillage and/or runoff of the small soil particles with relatively high RCs concentrations to areas outside the field.

### 3.1.4. Effect of manure application on soil chemical properties and grain RCs concentration

The soil ExK content was higher in the M1 plot than in the M0 plot at the buckwheat harvest time (Table 5). Other chemical properties did not differ between these two plots, although there were high values of exchangeable MgO, available P2O5, NO3-N, and humus in the M1 plot. Grain yield did not differ significantly between the two plots. The grain <sup>137</sup>Cs concentration in M1 was about half that in M0, although the values were not significantly different. We concluded that the relatively lower grain <sup>137</sup>Cs concentration in the M1 plot than in the M0 plot was due to the increased soil ExK content resulting from the application of cattle manure compost. In a paddy field in Fukushima Prefecture that had been physically decontaminated of RCs, the application of organic matter (rice straw and cattle manure compost) improved rice production and decreased the grain RCs concentration (Nishiwaki et al., 2017). We obtained similar results for buckwheat. Therefore, application of cattle manure compost is a practical

Table 5. Soil chemical properties, grain yield, and grain <sup>137</sup>Cs concentration in M0 and M1 plots.

	MO	M1
Soil		
рН	6.0	6.0
Exchangeable CaO (mg kg <sup>-1</sup> )	1570	1550
Exchangeable MgO (mg kg <sup>-1</sup> )	191	207
Exchangeable K <sub>2</sub> O (mg kg <sup>-1</sup> )	108	197
CEC (cmol <sub>c</sub> kg <sup>-1</sup> )	12.5	12.5
Available P <sub>2</sub> O <sub>5</sub> (mg kg <sup>-1</sup> )	194	233
$NH_4-N (mg kg^{-1})$	4.6	3.9
$NO_3-N (mg kg^{-1})$	2.9	3.2
$P_2O_5$ absorption coefficient (mg kg <sup>-1</sup> )	6840	6210
Humus (%)	5.5	5.7
Grain		
Yield (kg ha <sup>-1</sup> )	$706 \pm 14^{a}$	641 ± 71 <sup>ns b</sup>
<sup>137</sup> Cs (Bq kg <sup>-1</sup> )	16.8 ± 4.2	8.7 ± 0.1 <sup>ns</sup>

<sup>a</sup>Average  $\pm$  standard error (n = 2).

<sup>b</sup>ns, not significant.

countermeasure to increase the soil ExK content and decrease the crop RCs concentration in fields that have been physically decontaminated of RCs. This method is applicable for organic crop production, depending on the interest of the farmers.

### **3.2.** Buckwheat production by local farmers in RCs-decontaminated fields

**3.2.1.** Soil ExK content and grain RCs concentration Table 6 shows the soil <sup>137</sup>Cs concentrations and soil ExK contents before and during buckwheat cultivation in the fields of the KNBC in 2015. The average soil <sup>137</sup>Cs concentration in the three fields before cultivation was 231 Bq kg<sup>-1</sup>, lower than that in the experimental field where the on-site experiment was conducted (Section 3.1). The average soil ExK content before fertilization was <300 mg K<sub>2</sub>O kg<sup>-1</sup> in fields A and C. Out of five site measurements, the lowest values of soil ExK contents were 155, 153, and 75 mg K<sub>2</sub>O kg<sup>-1</sup> in fields A, B, and C,

**Table 6.** Soil <sup>137</sup>Cs concentration, soil ExK content, grain <sup>137</sup>Cs concentration, and <sup>137</sup>Cs transfer factor in the three fields (A, B, and C) cultivated by the KNBC.

. ,				
	А	В	С	
Soil <sup>137</sup> Cs (Bq kg <sup>-1</sup> )				
Before fertilization	$232 \pm 115^{a}$	316 ± 242	144 ± 42	
Sowing	389 ± 193	303 ± 282	117 ± 33	
Flowering	307 ± 115	474 ± 261	149 ± 18	
Maturity	254 ± 111	374 ± 210	164 ± 64	
Soil ExK content (mg $K_2O$ kg <sup>-1</sup> )				
Before fertilization	186 ± 12	300 ± 57	163 ± 39	
Sowing	559 ± 33	386 ± 40	506 ± 121	
Flowering	353 ± 23	303 ± 23	311 ± 23	
Maturity	363 ± 21	306 ± 76	304 ± 28	
Grain at maturity				
<sup>137</sup> Cs (Bq kg <sup>-1</sup> )	1.30 (1.62) <sup>b</sup>	4.68 (5.89)	2.32 (2.90)	
Transfer factor	0.0051	0.0127	0.0170	

<sup>a</sup> Average  $\pm$  standard error (n = 5).

<sup>b</sup> Values in parentheses are total concentrations of <sup>134</sup>Cs and <sup>137</sup>Cs.

respectively (data not shown). Application of potassium sulfate fertilizer to these fields before sowing increased the soil ExK content. The average soil ExK content was still >300 mg K<sub>2</sub>O kg<sup>-1</sup> at the buckwheat maturity stage in all the fields. The RCs (<sup>134</sup>Cs+ <sup>137</sup>Cs) concentrations in buckwheat grains harvested from these fields were under the standard limit (Table 6). The RCs concentrations in buckwheat grain were also under the standard limit in 2016 (1.69 Bq kg<sup>-1</sup>) and 2017 (<25 Bq kg<sup>-1</sup>). However, the TF of RCs in Fields B and C was not so low compared to that in the field where the on-site experiment was conducted, even though the soil ExK was mostly >300 mg  $K_2O$  kg<sup>-1</sup>. Several researchers such as Sawhney (1964), Nakao et al. (2008), and Mukai et al. (2016) have reported that RCs adsorb to clay minerals in the soil, and such adsorbed RCs are poorly absorbed by plants. Although further research is needed, differences in the soil clay mineral composition and the adsorption of RCs to soil may affect RCs uptake from soil by plants growing in contaminated fields.

Field B had a spot with a relatively low soil ExK content (137 mg  $K_2O$  kg<sup>-1</sup>) at buckwheat maturity in 2015; this may have been due to the erosion of dressed soil because field B was moderately sloping (Supplementary Figure 2). Wakabayashi et al. (2018) noted that there is a risk on runoff of RCs in soil by soil erosion from sloping fields, and suggested that cover crops should be grown to reduce soil erosion. Soil erosion may also reduce the soil ExK content, leading to increased RCs concentrations in crops. Therefore, preventing soil erosion is another important countermeasure.

### 3.2.2. Yields and significance of efforts by local farmers

In 2015, the total production of grain from fields A and C cultivated by the KNBC was 148 kg, and the yield was 269 kg ha<sup>-1</sup> (Table 7). The yield from field B was very low because of crop damage by wild boars. In 2016, the total grain production and yield were 240 kg and 240 kg ha<sup>-1</sup>, respectively. Some fields were affected by wild boars and water-lodging damage. In 2017, the fields were fenced with electric fences, and the total grain production and yield were 1070 kg and 713 kg ha<sup>-1</sup>, respectively. The buckwheat cultivated in these fields (Supplementary Figure 1c) featured at the harvest festival, which has been run by the KNBC at Kawamata each year since 2015 ('Fukushima-Minpo' on 8 December 2015, and 5 December 2017; 'Fukushima-Minyu' on 10 December, 7 December 2015, 2016, and 6 December 2017). This festival provides the opportunity for citizens affected by the evacuation to reconnect (Supplementary Figure 1d-f). From these aspects, demonstrative cultivation by farmers

 
 Table 7. Harvested area, total grain production, and grain yield in fields cultivated by KNBC.

	Harvested area (ha)	Total production (kg)	Yield (kg ha <sup>-1</sup> )
2015	0.55	148	269
2016	1.00	240	240
2017	1.50	1070	713

themselves and collaborations between farmers and researchers (Hoffmann et al., 2007) are effective methods to encourage better understanding of agricultural problems and the development of new techniques to share with farmers. The continuous support of the town office and the prefectural government are also contributing to demonstrative cultivation by farmers in fields physically decontaminated of RCs and the collaboration between farmers and researchers.

#### 4. Conclusion

This study revealed the status of fields physically decontaminated of RCs and the cautions for resuming agriculture in such fields. The main results were as follows:

- (1) Some RCs remained in the field even after physical decontamination. In the RCs-decontaminated fields, there were some 'hot spots' with high local RCs concentrations because of the uneven removal of radiation-contaminated surface soil and insufficient mixing of the original and dressed soils. Careful mixing of the soil and increasing the soil ExK content by adding potassium are important steps to decrease the concentration of RCs in buckwheat cultivated in decontaminated fields.
- (2) The soil fertility of RCs-decontaminated fields was relatively low compared with that of common fields. Nitrogen fertilizer had to be applied at double the conventional rate to support the growth of the first buckwheat crops in the RCsdecontaminated fields. Cattle manure also increased soil fertility and decreased the RCs concentration in buckwheat.
- (3) Buckwheat grain with radioactivity beneath the standard limit was produced by local farmers in the fields that had been physically decontaminated of RCs for 3 years. Resumption of agriculture with buckwheat cultivation is gradually progressing in Yamakiya District.

These results highlight that understanding the characteristics of the original and dressed soils in each area is important before relaunching agriculture. By adding appropriate inputs, farmers can produce crops with low RCs concentrations in physically decontaminated fields. Continuous field studies with the participation of local farmers could be a preface to relaunching crop production not only in Yamakiya District but also in other areas where evacuation orders have been lifted.

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